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EXPERIMENTAL INVESTIGATION OF THE THERMAL CONDUCTIVITY OF A BINARY Ar–Kr MIXTURE AT LOW TEMPERATURES

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New experimental results on the thermal conductivity of an Ar–Kr mixture in the 120-273°K temperature range are obtained.

There is a limited quantity of experimental results on the thermal conductivity of binary mixtures of monatomic gases in the T ≤ 273°K temperature range at atmospheric pressure. Wachsmuth [1] measured the thermal conductivity of an He–Ar mixture at atmospheric pressure and a temperature near 273°K (273.1°K), as did Rychkova and Golubev [2] within the limits 196.66-372.86°K and pressures from 0.1-29.4 mN/m^2. The authors of [2] obtained values of the thermal conductivity for four compositions of a helium–argon mixture in the low-temperature measurement range at 196.66°K and at pressures from 0.1-29.4 mN/m^2. Davidson and Music [3] measured the thermal conductivity of a helium–neon mixture at 273.1°K at atmospheric pressure, and Srivastava and Madan [4] performed the measurements for a neon–argon mixture.

We first investigated the thermal conductivity of an Ar–Kr mixture experimentally in the T = 120-273°K temperature range at atmospheric pressure for five argon concentrations: 25, 50, 75, 90.15, and 98.5%. High-purity argon with a 99.987% content of the main substance and 99.97% krypton were used in the tests. The measurements were performed on an apparatus employing the absolute hot-wire method [5].

Experimental results of the temperature dependences of the thermal conductivity of the mixture on composition are presented in Table 1; the following notation is used: Q_H is the quantity of heat transferred by conduction from the wire through the layer of mixture under investigation to the inner wall of the measuring tube; T_1 and T_2 are readings of the inner and outer resistance thermometers; Q is the total quantity of heat liberated by the heater; Q_R is the quantity of heat transferred by radiation from the heater to the wall of the measuring tube; Q_C is the quantity of heat transferred along the current supplying and potential conductors; λ is the thermal conductivity of the mixture; T_{av} is the reference temperature; and ΔT_{mix} is the temperature drop in the mixture layer under investigation.

Corrections for heat removal from the ends of the heater, radiation, and the temperature drop in the wall of the glass tube of the measuring cell were taken into account in processing the measurement results.

As measurements and calculations showed, corrections to the readings of the inner thermometer according to the outer for nonheating currents before each measurement (ΔT_{grad}), the temperature jump, eccentricity, and change in geometric size of the cell with temperature lie within the limits of experimental accuracy in the calculation of the thermal conductivity and hence were not introduced.

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TABLE 1. Experimental Results of the Determination of the Coefficient of Thermal Conductivity of an Ar—Kr Mixture

$Q\cdot10^5$, W	$Q_R\cdot10^5$, W	$Q_C\cdot10^5$, W	$Q_H\cdot10^5$, W	T_1, °K	T_2, °K	T_{av}, °K	$\lambda\cdot10^3$, W/(m·°K)	ΔT_{mix}, °K
				$X_1=0{,}985$				
672,3	0,07	17,4	654,8	99,30	93,57	96,44	6,49	5,72
414,7	0,07	10,1	404,5	111,65	108,45	110,05	7,19	3,19
547,8	0,1	12,8	534,9	119,57	115,58	117,58	7,62	3,98
643,8	0,2	14,8	628,8	124,50	119,91	122,21	7,79	4,58
646,7	0,2	14,9	631,6	125,23	120,70	122,97	7,93	4,52
685,0	0,2	19,5	665,3	127,60	122,86	125,23	7,98	4,73
690,4	0,3	14,6	675,5	139,49	135,09	137,29	8,73	4,39
935,9	0,5	19,5	915,9	154,46	149,01	151,74	9,55	5,44
1138,5	0,9	22,5	1115,1	171,67	165,67	168,67	10,56	5,99
1566,6	2,5	27,8	1536,3	213,76	207,00	210,38	12,91	6,75
2280,9	6,3	38,3	2236,3	253,53	245,01	249,27	14,92	8,50
3123,7	11,2	50,7	3061,8	276,23	265,37	270,80	16,03	10,83
				$X_1=0{,}9015$				
480,1	0,05	12,6	467,5	96,67	92,44	94,56	6,28	4,22
499,4	0,08	13,3	486,0	108,70	104,78	106,74	7,05	3,92
631,1	0,1	18,0	613,0	117,40	112,58	114,99	7,23	4,81
627,1	0,2	14,4	612,5	132,62	128,37	130,50	8,19	4,24
806,3	0,4	17,3	788,6	145,24	140,17	142,71	8,84	5,06
973,3	0,6	20,3	952,4	159,63	153,98	156,81	9,58	5,64
1072,0	0,9	21,2	1049,9	176,68	170,96	173,82	10,43	5,71
1352,9	2,0	25,6	1325,3	201,70	195,21	198,46	11,60	6,48
2103,9	4,7	37,4	2061,8	233,98	225,10	229,54	13,17	8,88
2472,1	8,0	41,7	2422,4	262,11	252,64	257,38	14,54	9,45
2457,9	9,1	52,6	2396,2	273,84	264,90	269,37	15,24	8,92
				$X_1=0{,}75$				
517,0	0,1	12,8	504,1	119,96	116,04	118,00	7,31	3,91
648,8	0,2	15,2	633,4	136,71	132,27	134,49	8,11	4,43
934,9	0,6	20,6	913,7	154,81	148,95	151,88	8,86	5,85
1248,9	1,0	26,5	1221,4	171,88	164,64	168,26	9,58	7,23
2205,1	2,8	44,6	2157,7	195,71	184,07	189,89	10,54	11,61
1777,4	2,9	35,6	1738,9	207,27	198,22	202,75	10,92	9,03
2213,6	4,8	41,8	2167,0	226,31	215,77	221,04	11,68	10,52
3516,3	9,3	65,9	3441,1	242,29	226,36	234,33	12,27	15,90
2511,2	8,3	45,0	2457,9	256,79	246,06	251,43	13,02	10,71
2932,7	12,5	50,9	2869,3	277,98	266,23	272,11	13,89	11,72
				$X_1=0{,}5$				
337,1	0,05	9,8	327,3	105,15	102,20	103,68	6,31	2,94
438,8	0,1	11,8	471,9	119,95	116,16	118,06	7,08	3,78
611,2	0,2	17,5	593,5	137,50	133,19	135,35	7,83	4,30
816,7	0,5	18,9	797,3	155,43	150,04	152,74	8,41	5,38
879,6	0,8	19,9	858,9	171,73	166,28	169,01	8,95	5,44
1154,0	1,6	24,3	1128,1	195,10	188,46	191,78	9,65	6,63
1459,5	2,9	34,6	1422,0	212,48	204,38	208,43	9,98	8,08
1564,0	4,4	30,6	1529,0	233,99	225,73	229,86	10,51	8,25
2273,6	8,7	47,2	2217,7	257,10	245,84	251,47	11,19	11,24
2363,1	10,3	41,8	2311,0	266,88	255,49	261,19	11,53	11,37
2284,5	11,3	42,7	2230,5	276,86	266,17	271,52	11,83	10,69
				$X_1=0{,}25$				
379,4	0 1	11,5	367,8	114,81	110,96	112,89	5,43	3,84
336,0	0,1	12,1	323,8	125,82	122,74	124,28	5,96	3,08
325,4	0,2	8,9	316,3	140,35	137,58	138,97	6,48	2,77
425,9	0,3	10,4	415,2	159,28	156,04	157,66	7,27	3,24
487,0	0,6	11,9	474,5	174,86	171,33	173,10	7,65	3,52
686,7	1,1	16,0	669,6	191,69	187,04	189,37	8,18	4,64
1012,9	2,4	22,4	988,1	214,70	208,26	211,48	8,72	6,43
1329,0	4,4	28,2	1296,4	236,53	228,45	232,49	9,11	8,07
1585,8	6,9	33,5	1545,4	253,93	244,56	249,25	9,36	9,36
1946,0	11,4	39,6	1895,0	279,13	268,52	273,83	10,15	10,59

No dependence of the effective values of the thermal conductivity λ on ΔT_{mix} is detected in the measurements, which indicates the absence of convection and is verified by a computation ($Gr \cdot Pr < 1000$).

Experimental data on the thermal conductivity represented in Table 1 are described by the polynomials

$$\begin{aligned}\lambda_{0.985} &= 0.1096 \cdot 10^{-2} + 0.5555 \cdot 10^{-4} T, \\ \lambda_{0.9015} &= 0.1502 \cdot 10^{-2} + 0.5066 \cdot 10^{-4} T, \\ \lambda_{0.75} &= 0.2436 \cdot 10^{-2} + 0.4208 \cdot 10^{-4} T, \\ \lambda_{0.5} &= -0.5216 \cdot 10^{-2} + 0.17687 \cdot 10^{-3} T - 0.7697 \cdot 10^{-6} T^2 + 0.1298 \cdot 10^{-8} T^3, \\ \lambda_{0.25} &= -0.4998 \cdot 10^{-2} + 0.1428 \cdot 10^{-3} T - 0.5403 \cdot 10^{-6} T^2 + 0.804 \cdot 10^{-9} T^3,\end{aligned} \tag{1}$$

where, for instance, $\lambda_{0.985}$ [W/(m•°K)] is the thermal conductivity of the mixture for a 0.985 molar concentration of argon.

Deviations of the experimental from the results calculated by means of (1) do not exceed ±1.5% and are +2% only for the mixture $X_1 = 0.9015$ at $T_{av} = 106.74$°K. The error analysis we made showed that it does not exceed 2.1%.

The measurement results were compared with corresponding theoretical values of λ calculated according to strict molecular-kinetic theory [6] by means of the formula

$$\lambda = 4 \frac{\begin{vmatrix} L_{11} & L_{12} & X_1 \\ L_{21} & L_{22} & X_2 \\ X_1 & X_2 & 0 \end{vmatrix}}{\begin{vmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{vmatrix}}, \tag{2}$$

where

$$L_{ii} = \frac{4X_i^2}{\lambda_i} - \frac{16T}{25p} \sum \frac{\left(\frac{15}{2} M_i^2 + \frac{25}{4} M_k^2 - 3M_k^2 B_{ik}^* + 4M_i M_k A_{ik}^*\right) X_i X_k}{(M_i + M_k)^2 D_{ik}},$$

$$L_{ij} = \frac{16T}{25p} \cdot \frac{X_i X_j M_i M_j}{(M_i + M_j)^2 D_{ij}} \left(\frac{55}{4} - 3B_{ij}^* - 4A_{ij}^*\right), \quad i \neq j,$$

$$\lambda_i = 0.08328 \frac{\sqrt{T/M_i}}{\sigma_i^2 \Omega^{(2,2)*}(T_i^*, \alpha_i)},$$

$$\frac{pD_{ij}}{T} = 0.026628 \frac{\sqrt{\frac{T(M_i + M_j)}{2M_i M_j}}}{\sigma_{ij}^2 \Omega_{ij}^{(1,1)*}(T_{ij}^*, \alpha_{ij})}.$$

Different representations of the intramolecular interaction were used to compute the thermal conductivity of the Ar–Kr mixture: the Lennard-Jones potential function (12–6) modified by the Buckingham potential (exp–6). The parameters of the potential functions are presented in Table 2, where ε_{12}/k and σ_{12} (the asterisk characterizes the interaction between inhomogeneous molecules) were calculated by using the following combination rules:

$$\frac{\varepsilon_{12}}{k} = \left(\frac{\varepsilon_1}{k} \cdot \frac{\varepsilon_2}{k}\right)^{\frac{1}{2}}, \quad \sigma_{12} = \frac{1}{2}(\sigma_1 + \sigma_2). \tag{3}$$

The results of the computations are presented in Table 3.

Comparing the theoretical results obtained with the experimental data showed that the most acceptable functions of intramolecular interaction to calculate the thermal conductivity

TABLE 2. Parameters of Interactions between Homogeneous and Inhomogeneous Molecules

Potential function	Variant	$\frac{\varepsilon_1}{k}$, °K	$\frac{\varepsilon_2}{k}$, °K	$\frac{\varepsilon_{12}}{k}$, °K	σ_1, Å	σ_2, Å	σ_{12}, Å	α_1	α_2	α_{12}	Reference
(12—6)	I	124,0	190,0	153,49*	3,418	3,61	3,514*	—	—	—	[6]
(12—6)	II	135,0	193,0	161 42*	3,36	3,57	3,465*	—	—	—	[7]
(12—6)	III	152,08	211,3	180,61	3,291	3,518	3,391	—	—	—	[8]
(exp—6)	I	123,2	158,3	143,0	3,866	4,056	3,947	14,0	12,3	13,3	[9]
(exp—6)	II	138,0	196,0	150,0	3,77	4,01	3,94	14,8	15,8	15,0	[7]

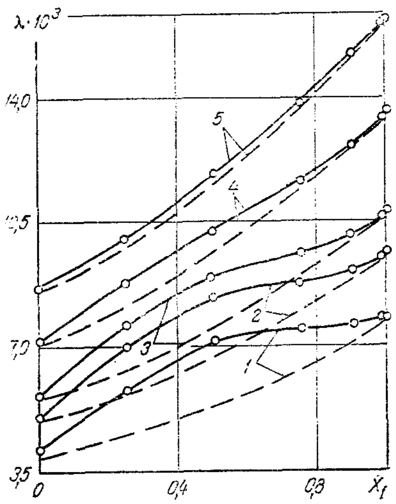

Fig. 1. Concentration dependence of the thermal conductivity of the Ar—Kr mixture: 1) T = 120°K; 2) 150°K; 3) 170°K; 4) 220°K; 5) 270°K. Points are experimental data; dashed curves are the theoretical values calculated by means of the Lennard-Jones (12—6) potential (potential parameters [6]).

of the Ar—Kr mixture in the temperature range under consideration are the Lennard-Jones potentials (12—6) (variants I and II) with potential parameters [6, 7] and the Buckingham potential (exp—6) with intramolecular interaction parameters [9]. However, it is difficult to assign preference to any of these potentials, since the discrepancy between the theoretical and experimental results for the very same concentrations of the light component in the mixture is approximately identical and increases with the reduction in temperature (see Table 3).

The concentration dependence of the thermal conductivity of an Ar—Kr mixture is presented in Fig. 1. The experimental curves were obtained by means of (1) and the theoretical curves by using the Lennard-Jones potential (12—6) (variant I) with potential parameters [6]. The theoretical data on the thermal conductivity of argon and krypton for T = 120 and 270°K, computed by using the Lennard-Jones potential (12—6) for the potential parameters [6], and the experimental results were taken from [5, 10].

It is seen from Fig. 1 that the concentration dependences of the thermal conductivity of the Ar—Kr mixture at temperatures below 220°K have an inflection point. The discrepancy between the theoretical and experimental results increases with the increase in concentration of the heavy component in the mixture and reaches 26% at T = 120°K for an equimolar mixture.

The experimental results on the temperature dependence of the thermal conductivity of this mixture do not exhibit the presence of a minimum for low krypton concentrations, as occurs in the case of measuring the thermodiffusion constant.

TABLE 3. Experimental and Calculation Results on the Thermal Conductivity of the Ar–Kr Mixture

T, °K	$\lambda_{exp}\cdot 10^3$	Potential (12–6), I		Potential (12–6), II		Potential (12–6), III		Potential (exp–6), I		Potential (exp–6), II	
		$\lambda_{calc}\cdot 10^3$	Δ, %	$\lambda_{calc}\cdot 10^3$	Δ, %	$\lambda_{calc}\cdot 10^3$	Δ, %	$\lambda_{calc}\cdot 10^3$	Δ, %	$\lambda_{calc}\cdot 10^3$	Δ, %
						$X_1 = 0{,}985$					
96,44	6,49	6,062	—6,6	6,055	—7,5	5,888	—9,3	6,033	—7,0	6,010	—7,4
110,05	7,19	6,934	—3,6	6,870	—4,5	6,731	—6,4	6,904	—4,0	6,859	—4,6
117,58	7,62	7,417	—2,7	7,346	—3,6	7,201	—5,5	7,377	—3,2	7,321	—3,9
122,21	7,79	7,738	—0,67	7,642	—1,9	7,493	—3,8	7,676	—1,5	7,615	—2,2
122,97	7,93	7,759	—2,2	7,690	—3,0	7,540	—4,9	7,726	—2,6	7,664	—3,4
125,23	7,98	7,902	—1,0	7,832	—1,9	7,679	—3,8	7,860	—1,5	7,806	—2,2
137,29	8,73	8,658	—0,80	8,593	—1,6	8,433	—3,4	8,593	—1,6	8,555	—2,0
151,74	9,55	9,549	—0,01	9,490	—0,63	9,332	—2,3	9,498	—0,54	9,411	—1,4
168,67	10,56	10,569	+0,10	10,527	—0,31	10,364	—1,9	10,514	—0,44	10,453	—1,0
210,38	12,91	12,953	+0,33	12,950	+0,31	12,833	—0,60	12,897	—0,10	12,851	—0,46
249,27	14,92	15,015	+0,64	15,068	+0,99	14,996	+0,51	14,975	+0,37	14,976	+0,38
270,80	16,03	16,093	+0,39	16,187	+0,98	16,156	+0,79	16,014	—0,10	16,079	+0,30
						$X_1 = 0{,}9015$					
94,56	6,28	5,592	—11,0	5,552	—11,6	5,452	—13,2	5,531	—11,9	5,499	—12,4
106,74	7,05	6,327	—10,2	6,281	—10,9	6,164	—12,6	6,255	—11,3	6,203	—12,0
114,99	7,23	6,817	—5,7	6,770	—6,4	6,643	—8,1	6,747	—6,7	6,680	—7,6
130,50	8,19	7,743	—5,5	7,692	—6,1	7,552	—7,7	7,646	—6,6	7,575	—7,5
142,71	8,84	8,455	—4,4	8,412	—4,8	8,266	—6,5	8,358	—5,5	8,267	—6,5
156,81	9,58	9,267	—3,3	9,231	—3,6	9,086	—5,2	9,154	—4,4	9,053	—5,5
173,82	10,43	10,223	—2,0	10,007	—4,1	10,062	—3,5	10,121	—3,0	10,000	—4,1
198,46	11,60	11,558	—0,36	11,559	—0,35	11,440	—1,4	11,451	—1,3	11,330	—2,3
229,54	13,17	13,163	—0,05	13,210	+0,30	13,116	—0,41	13,044	—0,96	12,932	—1,8
257,38	14,54	14,529	—0,08	14,606	+0,45	14,224	—2,2	14,386	—1,1	14,303	—1,6
269,37	15,24	15,097	—0,94	15,196	—0,29	15,164	—0,50	14,950	—1,9	14,898	—2,2
						$X_1 = 0{,}75$					
118,00	7,31	6,153	—15,8	6,247	—14,5	6,145	—15,9	6,186	—15,4	6,093	—16,6
134,49	8,11	7,143	—11,9	7,126	—12,1	7,008	—13,6	7,040	—13,2	6,932	—14,5
151,88	8,86	8,051	—9,1	8,040	—9,3	7,919	—10,6	7,936	—10,4	7,789	—12,1
168,26	9,58	8,885	—7,4	8,885	—7,4	8,766	—8,6	8,765	—8,5	8,598	—10,2
189 89	10,54	9,959	—5,5	9,976	—5,4	9,867	—6,4	9,821	—6,8	9,636	—8,6
202,75	10,92	10,577	—3,1	10,609	—2,8	10,506	—3,8	10,443	—4,4	10,235	—6,3
221,04	11,68	11,444	—2,0	11,486	—1,7	11,433	—2,1	11,298	—3,3	11,081	—5,1
234,33	12,27	12,049	—1,8	12,115	—1,3	12,041	—1,9	11,897	—3,0	11,670	—4,9
251,43	13,02	12,817	—1,6	12,895	—0,96	12,846	—1,3	12,652	—2,8	12,419	—4,6
272,11	13,89	13,712	—1,3	13,842	—0,35	13,792	—0,71	13,519	—2,7	13,300	—4,2
						$X_1 = 0{,}5$					
103,68	6,31	4,619	—26,8	4,629	—26,6	4,561	—27,7	4,612	—26,9	4,511	—28,5
118,06	7,08	5,264	—25,6	5,268	—25,6	5,193	—26,7	5,261	—25,7	5,122	—27,7
135,35	7,83	6,025	—23,1	6,042	—22,8	5,952	—23,9	6,032	—23,0	5,855	—25,2
152,74	8,41	6,792	—19,2	6,817	—18,9	6,718	—20,1	6,797	—19,2	6,574	—21,8
169,01	8,95	7,493	—16,3	7,526	—15,9	7,685	—14,1	7,509	—16,1	7,241	—19,1
191,78	9,65	8,461	—12,3	8,480	—12,1	8,410	—12,8	8,474	—12,1	8,164	—15,4
208,43	9,98	9,151	—8,3	9,180	—8,0	9,133	—8,5	9,159	—8,2	8,821	—11,6
229,86	10,51	10,004	—4,8	10,087	—4,0	10,011	—4,7	10,030	—4,6	9,642	—8,3
251,47	11,19	10,847	—3,1	10,942	—2,2	10,890	—2,7	10,867	—2,9	10,449	—6,6
261,19	11,53	11,217	—2,7	11,321	—1,8	11,277	—2,2	11,232	—2,6	10,802	—5,8
271,52	11,83	11,592	—2,0	11,707	—1,0	11,666	—1,4	11,612	—2,1	11,183	—5,7
						$X_1 = 0{,}25$					
112,89	5,43	4,231	—22,1	4,266	—21,4	4,205	—22,5	4,388	—19,2	4,198	—22,7
124,28	5,96	4,659	—21,8	4,697	—21,2	4,626	—22,3	4,841	—18,8	4,612	—22,6
138,97	6,48	5,212	—19,6	5,255	—18,9	5,175	—20,1	5,425	—16,3	5,148	—20,6
157,66	7,27	5,912	—18,7	5,965	—17,9	5,875	—19,2	6,158	—15,3	5,819	—20,0
173,10	7,65	6,484	—15,2	6,543	—14,5	6,455	—15,6	6,756	—11,7	6,732	—16,7
189,37	8,18	7,113	—13,0	7,138	—12,7	7,055	—13,7	7,383	—9,7	6,947	—15,1
211,48	8,72	7,855	—9,9	7,959	—8,7	7,869	—9,7	8,202	—5,9	7,710	—11,6
232,49	9,11	8,611	—5,5	8,710	—4,4	8,626	—5,3	8,971	—1,5	8,429	—7,5
249,25	9,36	9,186	—1,9	9,293	—0,72	9,218	—1,5	9,567	+2,2	8,978	—4,1
273,83	10,15	10,008	—1,4	10,130	—0,20	9,732	—4,1	10,406	+2,5	9,783	—3,6

Note. λ, W/(m·°K); $\Delta = [(\lambda_{calc} - \lambda_{exp})/\lambda_{exp}]\cdot 100$, %.

NOTATION

T, temperature, °K; p, pressure, N/m²; M_i, molecular weight of the i-th component of the mixture, kg/kmole; X_i, molar fraction of the i-th component of the mixture (the subscript 1 refers to the light component); λ_i, thermal conductivity of the i-th component of the mixture, W/(m·°K); D_{ii}, D_{ij}, self-diffusion factor of the i-th component of the mixture and mutual diffusion factor, m²/sec; ε, σ, parameters of the intramolecular interaction potential functions, °K, Å; $\Omega^{(l,s)*}(T^*)$, collision integrals; A*, B*, quantities expressing the collision integrals.

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RELATIONSHIPS BETWEEN COMPLEXES OF THERMOPHYSICAL PROPERTIES AND COMPRESSIBILITIES OF FLUIDS

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Based on experimental data for water and toluene, relationships between complexes of thermophysical quantities at a given temperature and pressure and between them and compressibility complexes are established.

It was established [1-11] that the thermal conductivity, dynamic viscosity, thermal activity, isobaric heat capacity, sound velocity, and Prandtl number for fluids are expressed by equations similar to an equation of state resembling the equation of state for actual gases in virial form which include a combination of only two of the so-called virial coefficients depending on the type of fluid and the temperature. These equations for water and six aromatic hydrocarbons (benzene, toluene, ethylbenzene, m-, p-, and o-xylols) have the following form:

for water

$$\frac{pv}{RT} = 1 + B\rho + E\rho^4, \quad (1)$$

$$\frac{\lambda}{\lambda_s'} = 1 + B_\lambda\rho + E_\lambda\rho^4, \quad (2)$$

$$\frac{\eta}{\eta_s'} = 1 + B_\eta\rho + E_\eta\rho^4, \quad (3)$$

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